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***Ab Initio* Description of Electronic Transport**

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Ab initio calculations of electronic transport became an important tool for the interpretation of quantum transport experiments on the atomic scale. Different theoretical concepts used in these kind of calculations are presented. The Landauer equation is introduced, its basic assumptions and the corresponding limits of applicability are discussed and compared to a calculation of the tunneling current based on perturbation theory. We also give a glimpse on the techniques used to treat open systems and systems out of equilibrium by means of the Green function approach.

1 Introduction

Calculating the electronic current flowing in a system due to some bias voltage is one of the most difficult problems in theoretical solid state physics. Different levels of approximations have been applied to the problem and theoretical models for many different aspects of the problem have been discussed. Among these, one of the most well known methods to treat electronic transport in solids is based on the Boltzmann formalism, which describes the time-dependent change in the electronic distribution function due to the applied field \vec{E} . It is a classical approximation as it considers the electrons to be moving freely between individual scattering events. Interference effects due to the quantum nature of the electrons are neglected. On the other hand, the very fact that the electrons move in a solid without scattering at every atomic site, i.e. the possibility to describe the electrons as (quasi) particles moving freely around is of course a quantum mechanical phenomenon. Very often, the Boltzmann equation is used in the so-called relaxation-time approximation, assuming a simple linear dependence of the scattering rate of the conducting electrons on the change Δf of the distribution function f that can be expressed in terms of a characteristic scattering time τ or alternatively in terms of a characteristic average length $L = \tau v$ between two scattering events. In this approximation, the Boltzmann formula takes the form

$$\frac{\partial}{\partial t} f + \vec{v} \frac{\partial}{\partial r} f - e \vec{E} \frac{1}{\hbar} \frac{\partial}{\partial k} f = -\frac{1}{\tau} \Delta f . \quad (1)$$

The average length between two such scattering events, the so-called mean-free path is the basic input parameter in the Boltzmann formula (Figure 1). One can derive approximations for different contributions to such scattering events and e.g. find the low-temperature dependence of the resistance of simple metals due to electron-phonon scattering as given in the famous Bloch T^5 law.

The approximations used in the Boltzmann approach restrict its applicability to the case in which the sample dimensions are much larger than the mean-free path and at the same time the scattering events can be viewed as independent from each other, i.e. quantum interference effects can be neglected. Such quantum interference can lead to a drastic variation of the resistance of a sample in an effect known as Anderson localization.

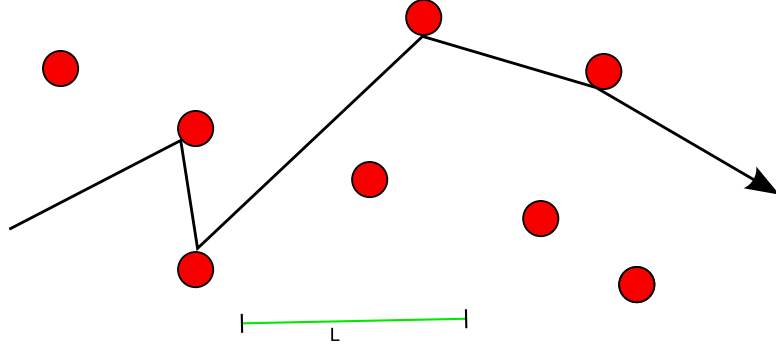


Figure 1. Scattering of electrons traveling in a solid. The average distance between such scattering events is the mean-free path L .

A more basic approach to the problem of electric conductance uses the idea of treating the current as a response of the quantum system to the applied electric field. In its most rigorous formulation this idea can be used in the linear response limit in which one assumes that the current depends linearly on the field. Using the corresponding time-dependent quantum mechanical formulation one obtains the so-called Kubo formalism which can be used to obtain the conductance of the system. While this approach is very general and would allow to include all kinds of scattering, it is not easy to apply it to realistic systems.

In the following, we will concentrate on calculating the electronic transport from a quantum mechanical point of view. In particular, we will consider systems with typical dimensions much smaller than the mean-free path due to scattering at structural impurities, by interface roughness, by phonons, magnons or other temperature dependent excitations present in real experiments. In the nanosize setups we consider, the resistance is due to the scattering of the electrons on the potential change in some scattering volume. A typ-

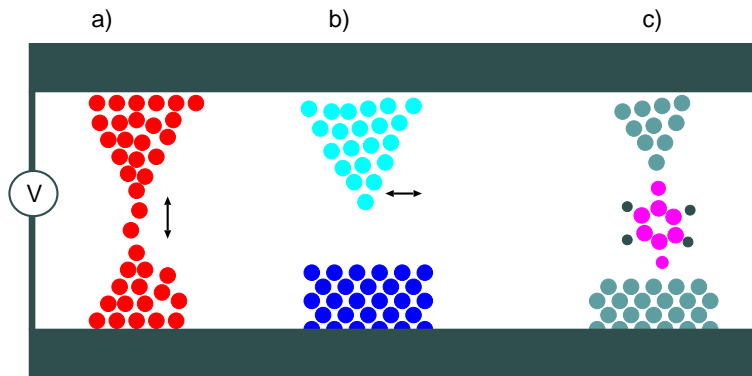


Figure 2. Different experimental setups used in nanoscale transport experiments. a) Metallic nanowires which might have different length, b) Scanning tunneling microscopy with a tip scanning the surface, c) Transport through single molecules.

ical experimental setup of such a system would be a break-junction with an atomic wire suspended between the two electrodes or a tunneling setup like a scanning tunneling microscope (STM) (Figure 2a,b)). The same approximation is also frequently applied to electron transport through single molecules (Figure 2c)) even though one could expect that scattering due to molecular vibrations will be important here.

This lecture will cover two main subjects. First, we will discuss the basic models to describe electron transport in a single-particle picture. This part will cover the famous Landauer approach to ballistic transport (Section 2.1) as well as a more specialized approach to the tunneling problem – Bardeen’s approach based on perturbation theory (Section 2.3). After introducing these models, we will focus on the specific problems one faces when trying to apply these ideas using the density functional theory. In particular we will discuss how to deal with semi-infinite leads and open systems without periodicity (Section 3). Our discussion will close with a short summary of the basic ideas applied to treat a non-equilibrium situation with a finite applied bias voltage to a transport junction (Section 3.1).

2 Single-Particle View on Quantum Transport

Even when restricting the theoretical description to the electron system only, the transport process is actually a complicated many electron problem of a system in non-equilibrium. On the macroscopic scale one can already define some of the different quantities describing the system like current and charge density or the applied electric field. However, it is very difficult to track these quantities down to the microscopic scale due to the complicated thermodynamically averaging taking place. In the following we will only deal with the very restricted subset of phenomena that are due to quantum mechanical nature of the electrons and we will therefore consider systems which can be described by pure wavefunctions without any statistical averaging. Still, in this picture one would have to describe the electron transport by the time-dependent many-body wavefunction $\Psi(t)$. For example, one could consider the probability $P_{i,f}$ of the system changing its state from some initial multi-electron state Ψ_i into a final multi-electron state Ψ_f where the two states differ with respect to their charge distribution. Thus, this approach makes it necessary to calculate the many particle time-dependent wavefunctions of the entire system. This is a very difficult task which cannot be solved in general.

To overcome this fundamental obstacle we will switch to the single-electron picture of electron transport. Similar to the replacement of the many particle problem of determining the ground state of a many-body quantum mechanical system in density functional theory by a single-particle Kohn-Sham formalism we will treat the electronic transport as due to the transport of many independent single-electrons. Furthermore, we will assume that the single-particle states in the Kohn-Sham formulation actually describe these independent single-electrons. Of course, there can be no hope that this very simplistic model actually is able to catch all the essential physics of the transport process. However, in analogy to standard band structure calculations of solids in equilibrium in which in many cases the Kohn-Sham single particle eigenvalues and eigenstates can be successfully interpreted as the elementary excitations of the systems we will apply the same procedure to the electronic transport and assume that the effects of the atomic arrangement, of the electronic (self-consistent) charge density and single-particle potential on the current can actually be modeled by this approach.

Many effects restrict the validity of the single-electron approach. Most obvious might be electron-electron scattering effects of different conducting electrons, but also interactions with the lattice beyond the static approximation, i.e. electron-phonon scattering, screening and charging effects or many particle interactions in magnetic systems might limit the validity of the single-electron picture. Only if these processes are sufficiently weak, one can hope that the single-particle approximations in terms of the Kohn-Sham states will provide reasonable results. This corresponds to the limit discussed in the introduction in which the mean-free path is much larger than the system size. In the single-particle picture the description in terms of the Kohn-Sham wavefunction will hold only on length scales shorter than this length scale, since these processes not included in the model will lead to the scattering destroying the phase coherence between the single-electron states involved.

One should note, that in many cases this description of the electron current in terms of single-particle physics is a completely inappropriate point of view. As soon as quantum many-body effects come into play, qualitatively different phenomena can be observed. Examples of such effects are the Kondo-effect in which a two level quantum scatterer embedded in a metallic environment leads to conductance abnormalities at low temperatures or correlation effects like the Coulomb blockade which are not reproduced in standard density functional theory treatments.

Obviously our description does not include any processes by which the single particle energy of the states carrying the current is changed. Hence only elastic transport can be described. This kind of quantum transport with only elastic scattering included is frequently called ballistic transport in mesoscopic physics as in many aspects the electrons behave like classical particles moving in a “billiard” like fashion. However, one should be aware of underlying quantum mechanical picture of single-electron states by which the electrons are described. These single-particle states describe the movement of the electrons between scattering events which scatter them from one single particle state into another.

2.1 The Landauer Approach

Landauer¹ proposed a theory of the transport process which is well adapted to describe the ballistic transport. A very intuitive and simple derivation will be presented here. The Landauer equation can also be derived more rigorously starting from linear response theory. In the Landauer approach to transport one considers the region Ω in which the electrons travel ballistically to be attached to two reservoirs L and R (see Figure 3).

The conductance Γ of the region Ω is defined by the current I_{LR} divided by the potential difference between the two reservoirs. The current I_{LR} on the other hand is given by the current due to all electrons traveling from L to R minus the current due to the electrons traveling vice versa

$$I_{LR} = I_{L \rightarrow R} - I_{R \rightarrow L} . \quad (2)$$

To arrive at an equation for these currents, one can start with a simple one-dimensional model. The current from the left to the right is determined by all electrons leaving the left reservoir, entering the scattering region Ω , and leave this scattering region by passing into the right reservoir. If one now assumes a very simple picture of the region Ω in which its electronic structure is described by single band in which states with $k > 0$ propagate from

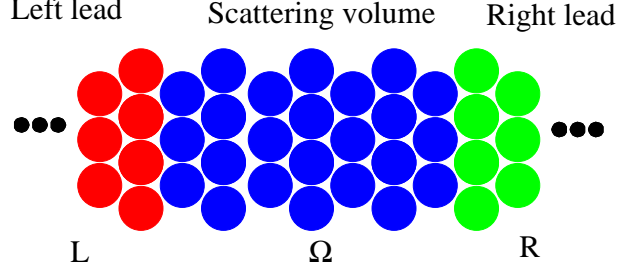


Figure 3. Basic setup of the Landauer approach to transport. The leads act as reservoirs and are assumed to be in thermal equilibrium.

the left to the right the current is given by an integral over all states with $k > 0$ up to the Fermi wave-vector k_F

$$I_{L \rightarrow R} = \int_0^{k_F} e v(k) dk, \quad (3)$$

where v denotes the group velocity of the state. Since

$$v = \frac{1}{\hbar} \frac{\partial E}{\partial k} \quad (4)$$

and converting the integral over k into an energy integration using the density of states $n(E)$,

$$\begin{aligned} I_{L \rightarrow R} &= \int_0^{\mu_L} \frac{e}{\hbar} \frac{\partial E}{\partial k} n(E) dE \\ &= \int_0^{\mu_L} \frac{e}{\hbar} \frac{\partial E}{\partial k} \frac{1}{\partial E / \partial k} \frac{1}{2\pi} dE \\ &= \int_0^{\mu_L} \frac{e}{\hbar} dE = \frac{e}{\hbar} \mu_L, \end{aligned} \quad (5)$$

where the energy integration has to be performed over all energies up to the Fermi energy (the chemical potential) of the left reservoir. This can be understood from the requirement that the electrons were assumed to be incoming from the left and therefore must be occupied in the reservoir.

Using the same derivation for the states incoming from the right reservoir one obtains

$$I_{LR} = \frac{e}{\hbar} (\mu_L - \mu_R). \quad (6)$$

Identifying the difference in the chemical potentials μ_L and μ_R with the applied voltage $eV = (\mu_L - \mu_R)$ one obtains the following interesting equation for the conductance

$$\Gamma = \frac{I_{LR}}{V} = \frac{e^2}{\hbar}. \quad (7)$$

This equation is truly remarkable since it states that each conducting band contributes the same to the conductance. Irrespectively of the density of states or the group velocity of the conducting states the conductance is always given by the fundamental quantum of conductance $\frac{e^2}{h}$. Indeed, as Eq. (5) shows, states with a low velocity and therefore a low current $j = ev$ are compensated by their higher density of states such that the conductance remains constant.

In the case of multiple bands, the derivation has to be modified by the inclusion of an extra sum over the different bands. Therefore in the general case of N conducting bands one obtains

$$\Gamma = \frac{e^2}{h} N . \quad (8)$$

The different „bands“ in this discussions are usually called „channels“. The argumentation presented so far did not care about the proper definition of these channels. These were simply assumed to form some kind of „band“ within Ω described by the usual formalism of a wave-vector k and a dispersion relation $E(k)$. Strictly speaking, since the system is not periodic, one cannot speak of Bloch states with some wave-vector having a component k in the direction of the current.

Since a key point in the discussion was the preparation of a state traveling from within the reservoirs through the region Ω one should clarify this idea. For such a state traveling to the right, one might assume the typical scattering problem. Within the left reservoir one considers a wavefunction being a Bloch state propagating towards the region Ω . „Propagate towards“ in this context should be understood as a state having a current flowing towards Ω . Within the reservoirs the resulting scattering state can be written in terms of reflected ψ_r and transmitted ψ_t states which are all solutions of the bulk Schrödinger equation in the reservoirs with the same energy as the incoming state ψ_{in} . The k values of these transmitted and reflected states have to be chosen such that the states „propagate away“ from Ω .

$$\psi(\vec{r}) = \begin{cases} \psi_{in}(\vec{r}) + \sum_n r_{in,n} \psi_r^n(\vec{r}) & \vec{r} \text{ in left reservoir} \\ \sum_{n'} t_{in,n'} \psi_t^{n'}(\vec{r}) & \vec{r} \text{ in right reservoir} . \end{cases} \quad (9)$$

Here the summations can be considered to be performed over all reflected Bloch states or all transmitted Bloch states. In principle, also states decaying away from the interfaces into the reservoirs must be included in this expansion. However, since these do not carry any current and by shifting the interface far enough into the reservoirs one can eliminate these decaying states.

Looking back to the derivation of the Landauer formula, an important change has to be made. While in Eq. (3) and Eq. (4) the summation over the incoming states and the evaluation of the current from their group velocities were all performed within the same single band picture, now one has to distinguish more carefully. The k integration in Eq. (3) has to be performed over the „in“ label of the expansion in Eq. (9). The sum over the velocities on the other hand is best performed in the right electrode. This is possible since current is conserved and can be very easily be done if all transmitted states and the incoming state are normalized to carry unit current. Using the orthogonality of the Bloch states one can perform the same steps as in Eq. (2) to (6) again to derive the more general Landauer equation

for ballistic transport in the presence of some scattering of the incoming electrons,

$$\Gamma = \frac{e^2}{h} \sum |t_{ij}|^2, \quad (10)$$

where i, j label the Bloch states in the reservoirs traveling from the left to the right.

Eq. (10) allows a simple interpretation of the transport in terms of the underlying quantum mechanical property of the transmission probability $P_{ij} = |t_{ij}|^2$ of an electron from the incoming Bloch state i into the transmitted Bloch state j . This interpretation makes the requirement of normalizing the incoming and transmitted Bloch states to unit current very clear, since in this normalization the direct interpretation of this probability is reasonably well defined and Eq. (10) can be seen as a simple generalization of Eq. (8).

2.2 Interpretation of the Landauer Formula

The Landauer formula (Eq. (10)) was the source of some confusion for quite some time after its first formulation^{2,3}. The most striking feature of the equation might be its limit for a perfectly transmitting region, i.e. for a region with $P_{ij} = |t_{ij}|^2 = 1$ for some set of i, j . For example if one would consider a perfect bulk crystal sandwiched between reservoirs of the same bulk material the expansion of Eq. (9) would collapse to

$$\psi(\vec{r}) = \begin{cases} \psi_{\text{in}}(\vec{r}) + \sum 0\psi_r & \vec{r} \text{ in left reservoir} \\ 1 \psi_t = \psi_{\text{in}}(\vec{r}) & \vec{r} \text{ in right reservoir} \end{cases}, \quad (11)$$

and one would rediscover Eq. (8) with N denoting the number of incoming Bloch states. At first glance, this means that the Landauer equation predicts a limited conductance of a system without any de-coherent scattering, i.e. of a perfect bulk crystal. In the same way the Landauer equation would also give a finite conductivity of a free electron gas. In this case a question which can always be asked only becomes more obvious to ask: How can a region with ballistic transport, i.e. without any dissipative processes, have a finite conductance? Since there is a voltage drop over the region and a current is flowing, some energy must dissipate. The key to the answer to this question lies in the definition of the reservoirs which were assumed to be in thermal equilibrium with some chemical potential μ attached to them. This is only possible, if there are actually dissipative processes in the reservoirs leading to the „thermalization“ of the „hot“ electrons being transferred across the region of ballistic transport.

The surprising result of a finite conductance in the case of a perfect crystal can now be interpreted in different ways. Either the setup described was not correct, since the reservoirs could not remain in thermal equilibrium and being perfect crystals like the region of ballistic transport at the same time or, which is actually very much the same, no finite voltage can be applied across such a system. The finite conductance of such a system with perfect ballistic transmission can now be interpreted as due the finite resistance at the interface between the reservoir and the ballistic region. This is also called the Sharvin-resistance of the system.

Another point to mention in the discussion of the physical significance of the Landauer equation is its formulation in terms of a two-terminal device. Both the current and the voltage drop are defined between the same two reservoirs. In many experiments, especially in mesoscopic physics a four point measurement is performed in which the current is driven between electrodes different than those between the voltage drop is measured.

Büttiker² presented a generalization of the Landauer equation to these multi-terminal cases. While this approach is very appropriate for mesoscopic physics, on the atomic scale multi-terminal arrangements are not the typical experimental arrangements and thus Eq. (10) will be sufficient. As depicted in Figure 4, additional resistances present in the current circuit are frequently eliminated in a four-point measurement, in which two additional potential probes are attached close to the scattering volume. However, for scattering volumes on the atomic scale, these geometries are not appropriate and thus we will restrict ourselves to simple two point geometries.

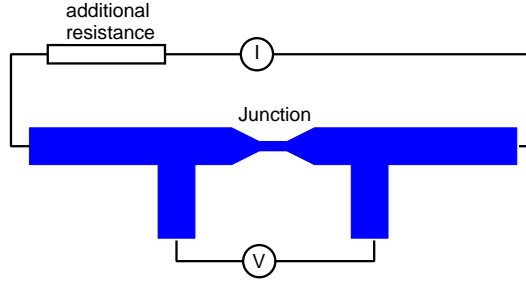


Figure 4. Four point geometry used experimentally to determine the voltage drop across a junction without including additional resistances present in the current circuit.

We will come back to this point when discussing the non-equilibrium transport later in this manuscript.

While the Landauer equation is valid in many cases reaching from systems with high conductivity to systems in the tunneling regime, one has to be careful in its application in some cases. Only states in which the incoming and transmitted waves can be described by Bloch states contribute to the tunneling current. This excludes any state which is localized within the region of ballistic transport to contribute. This corresponds to the fact that these states do not carry any current within the simple one-electron picture of transport chosen. In reality, there exist processes beyond this picture which lead to some coupling of these localized states to the otherwise orthogonal Bloch states in the reservoirs. For example the many-body electron-electron interaction, electron-phonon scattering, or structural defects not included in the description can provide such a coupling. Thus, while the Landauer approach will be correct for cases of high transmission through Bloch states, one could imagine that in the limit of a very low transmission probability other processes of transport across the ballistic region become important. In the one-electron picture these processes could be thought of as the transition of an electron from the reservoirs into some localized state of the reservoir, the transition of the electron from one side of the reservoir to the other and then the transition of the electron into a state of the other reservoir. The validity of the Landauer model is now limited by the transmission probability between the reservoir states and the localized state. If this probability becomes comparable to the probabilities $P_{ij} = |t_{ij}|^2$ the Landauer equation breaks down.

On the other hand, one can of course treat the other limit in which the transition probability between the two sides of the reservoir becomes very small and the details of the scattering processes needed to couple the states can be neglected. This limit can be suc-

cessfully described by theories for the quantum mechanical tunneling process.

2.3 The Bardeen Approach to Tunneling

The following description of the tunneling process is based on Bardeen's approach to tunneling which essentially applies time-dependent perturbation theory to the problem. Figure 5 shows the tunneling setup used in this approach. Two semi-infinite crystals are separated by a barrier region, which will be assumed to be a vacuum barrier for simplicity. If this vacuum barrier is sufficiently high and wide one can think the total setup to consist of two independent systems: one at the left (L) and one at the right (R) side.

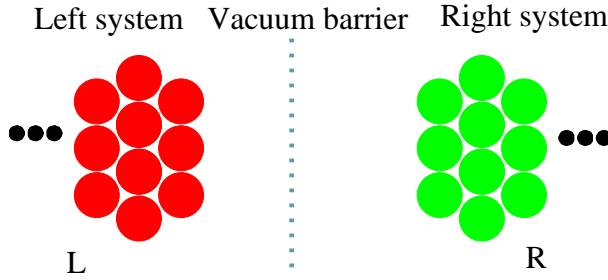


Figure 5. Tunneling setup used in Bardeen's approach to transport. The two semi-infinite crystals at the left and the right are separated by a vacuum barrier.

This total separation of the systems leads to two independent Schrödinger equations for the two sides

$$\begin{aligned} (T + U_L)\psi_L &= \epsilon_L\psi_L \\ (T + U_R)\psi_R &= \epsilon_R\psi_R \end{aligned} \quad (12)$$

where T denotes the operator of the kinetic energy of a single electron and U_L and U_R are the potentials of the left and right system respectively. The single-particle wavefunction $\psi(t)$ of the entire setup is determined by the total Hamiltonian $H = T + U_L + U_R$.

Now one can apply time-dependent perturbation theory to describe the tunneling of an electron across the vacuum barrier. Tunneling from the left to the right is assumed, the case of an electron tunneling vice versa may be treated completely analogously. The initial state of the tunneling process is localized in the left system. Therefore, there exists an eigenstate ψ_L^μ with $|\psi(t \rightarrow -\infty)\rangle = |\psi_L^\mu\rangle$. The time-dependence of the state $|\Psi(t)\rangle$ is governed by the Hamiltonian of the whole system.

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = H |\psi(t)\rangle . \quad (13)$$

The tunneling probability is given by the overlap of this time-dependent wavefunction with a wavefunction $|\psi_R^\nu\rangle$ of the right system. Multiplying Eq. (13) from the left with $\langle\psi_R^\nu|$ leads to

$$\langle\psi_R^\nu| \left(i\hbar \frac{\partial}{\partial t} \right) |\psi(t)\rangle = \langle\psi_R^\nu| H |\psi(t)\rangle . \quad (14)$$

Using the Schrödinger equation for the left state one obtains

$$i\hbar \frac{\partial}{\partial t} \langle \psi_R^\nu | \psi \rangle = \langle \psi_R^\nu | H | \psi \rangle - \langle \psi_R^\nu | H_R | \psi \rangle = \langle \psi_R^\nu | U_L | \psi \rangle \quad (15)$$

Substituting $|\psi(t \rightarrow -\infty)\rangle = |\psi_L^\mu\rangle$ for $|\psi\rangle$ at the right hand side of Eq. (15) leads to first order perturbation theory

$$i\hbar \frac{\partial}{\partial t} \langle \psi_R^\nu | \psi \rangle = \langle \psi_R^\nu | U_L | \psi_L^\mu \rangle. \quad (16)$$

Even though this equation looks familiar one has to emphasize that this is not a result obtained by standard time-dependent perturbation theory. The states $|\psi_L\rangle$ and $|\psi_R\rangle$ are eigenstates of the Hamiltonians H_L and H_R respectively. Therefore, they do not form a complete orthogonal basis of the eigenspace of the total Hamiltonian $H = T + U_L + U_R$ and the matrix elements at the left side of Eq. (16) are not sufficient to determine the total time-dependence of $|\psi\rangle$. This is a basic weakness of Bardeen's approach. However many applications⁴⁻⁷ of this formalism have shown that Bardeen's approximation produces reliable results for systems which are well separated, i.e. systems where the overlap of the two wavefunctions ψ_R and ψ_L is small.

Since the potential U_L is not small in the left region, the question arises whether one is allowed to use perturbation theory at all. However, it can be seen from Eq. (16) that the quantity which in fact determines the strength of the perturbation of the initial state is $\langle \psi_R^\nu | U_L | \psi_L^\mu \rangle$. Since the final wavefunction $|\psi_R\rangle$ is localized in the right region in which the left potential U_L is very weak this perturbation might still be regarded as a small perturbation and thus time-dependent perturbation will lead to reasonable results.

By separating the time-dependence of the states $|\psi_L^\mu\rangle = e^{i\epsilon_\mu t} |\Psi_L^\mu\rangle$ and $|\psi_R^\nu\rangle = e^{i\epsilon_\nu t} |\Psi_R^\nu\rangle$, integrating Eq. (16) and performing the limit $t \rightarrow \infty$, one obtains an expression for the tunneling probability per time interval

$$P_{\mu\nu}^{LR} = \lim_{t \rightarrow \infty} \frac{1}{t} \frac{1}{\hbar^2} \int_0^t |\langle \psi_R^\nu | U_L | \psi_L^\mu \rangle|^2 \quad (17)$$

$$= \lim_{t \rightarrow \infty} \frac{4}{\hbar} \frac{\sin^2\left(\frac{\epsilon_\nu - \epsilon_\mu}{2\hbar} t\right)}{(\epsilon_\nu - \epsilon_\mu)^2 t} |M_{\mu\nu}^{LR}|^2, \quad (18)$$

where the matrix element $M_{\mu\nu}^{LR}$ is given by the stationary-state matrix element of the potential

$$M_{\mu\nu}^{LR} = \langle \psi_R^\nu | U_L | \psi_L^\mu \rangle. \quad (19)$$

Assuming a continuous range of energy levels ϵ_μ (or ϵ_ν) the limit of Eq. (18) can be evaluated directly⁸. One obtains

$$P_{\mu\nu}^{LR} = \frac{2\pi}{\hbar} \delta(\epsilon_\nu - \epsilon_\mu) |M_{\mu\nu}^{LR}|^2. \quad (20)$$

This result is similar to the well known 'Golden Rule' Fermi obtained for standard time-dependent perturbation theory. It describes elastic tunneling with energy $\epsilon_\nu = \epsilon_\mu$ only. Formally this condition is taken care of by the δ -function in Eq. (20).

To evaluate this matrix element one can introduce an additional approximation. He assumed the potential U_L to be zero in the right region of space. Similar the right potential

should be zero in the left region. More formal one assumes a separation surface S which separates the regions in which the two potentials differ from zero. This can be written down by the condition $U_L U_R = 0$ for any point in space. Figure 6 shows the setup as used in this additional approximation.

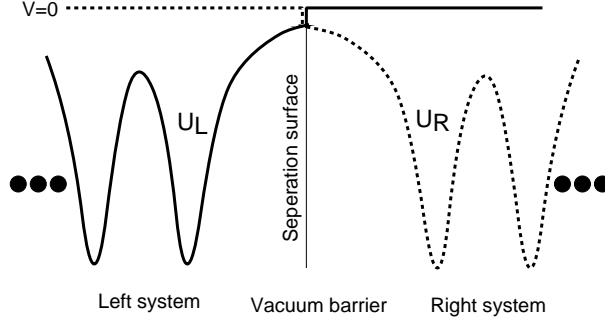


Figure 6. Potential used in the Bardeen approach to tunneling. The left (right) potential U_L (U_R) is then assumed to be zero in the right (left) region.

Of course, this approximation will become better if the potentials U_L and U_R are reasonably small at and beyond the separation surface. This will be the case if the separation surface is located far out in the vacuum.

Using the Schrödinger equation for the left wavefunction and having in mind that the potential U_L is zero in the right space one can now rewrite the matrix element as an integral over the left region only

$$M_{\mu\nu}^{LR} = \int_L \Psi_R^\nu(\vec{r})^* \left(\epsilon_\mu + \frac{\hbar^2}{2m} \vec{\nabla}^2 \right) \Psi_L^\mu(\vec{r}) dV \quad (21)$$

which can be written in a more symmetric form

$$\begin{aligned} M_{\mu\nu}^{RL} &= \int_L \left\{ \Psi_R^\nu(\vec{r})^* \epsilon_\nu \Psi_L^\mu(\vec{r}) + \Psi_R^\nu(\vec{r})^* \frac{\hbar^2}{2m} \vec{\nabla}^2 \Psi_L^\mu(\vec{r}) \right\} dV \\ &= \int_L \left\{ \Psi_R^\nu(\vec{r})^* \overleftarrow{(T + U_R)} \Psi_L^\mu(\vec{r}) + \Psi_R^\nu(\vec{r})^* \frac{\hbar^2}{2m} \vec{\nabla}^2 \Psi_L^\mu(\vec{r}) \right\} dV \\ &= -\frac{\hbar^2}{2m} \int_L \left\{ \Psi_L^\mu(\vec{r}) \vec{\nabla}^2 \Psi_R^\nu(\vec{r})^* - \Psi_R^\nu(\vec{r})^* \vec{\nabla}^2 \Psi_L^\mu(\vec{r}) \right\} dV \end{aligned} \quad (22)$$

In these transformations in the first step the eigenvalue ϵ_μ was substituted by ϵ_ν because energy conservation requires the calculation of matrix elements with $\epsilon_\mu = \epsilon_\nu$ only. In the second step the Schrödinger equation for the right state was used (the arrow indicates the wavefunction the operators acts on). The integration area is the left region. Since the potential U_R is assumed to be zero in this region, it was dropped in the last step. Using Greens theorem and the boundary condition that the right wavefunction is zero at infinite distance from the separation surface this integral can be transformed into an integral over

the separation surface

$$M_{\mu\nu}^{LR} = -\frac{\hbar^2}{2m} \int_S \left(\Psi_L^\mu(\vec{r}) \vec{\nabla} \Psi_R^\nu(\vec{r})^* - \Psi_R^\nu(\vec{r})^* \vec{\nabla} \Psi_L^\mu(\vec{r}) \right) dS. \quad (23)$$

So far only an expression for the probability of the transition of an electron from a left state into a right state was obtained.

Slightly modifying Eq. (20) this probability can be written as

$$P_{\mu\nu}^{LR} = \frac{2\pi}{\hbar} \delta(\epsilon_\mu^L - \epsilon_\nu^R - eV) |M_{\mu\nu}^{LR}|^2, \quad (24)$$

where the additional term eV is introduced to account for the bias voltage V applied between the two sides. To calculate the tunneling current one has to sum over all different possible left and right states and one has to keep in mind that the electrons might tunnel from the left to the right as well as vice versa. The total current therefore is given by

$$\begin{aligned} I &= I^{L \rightarrow R} - I^{R \rightarrow L} \\ &= e \sum_{\mu\nu} f(\epsilon_\mu)(1 - f(\epsilon_\nu + eV)) P_{\mu\nu}^{LR} - e \sum_{\mu\nu} (1 - f(\epsilon_\mu)) f(\epsilon_\nu + eV) P_{\nu\mu}^{RL} \\ &= e \sum_{\mu\nu} (f(\epsilon_\mu) - f(\epsilon_\nu + eV)) P_{\mu\nu}^{LR} \end{aligned} \quad (25)$$

where $f(\epsilon)$ denotes the Fermi-distribution function which is introduced to ensure that only tunneling from occupied to unoccupied states can occur. In Eq. (25) the symmetry of the tunneling probability $P_{\mu\nu}^{LR} = P_{\nu\mu}^{RL}$ which can easily be deduced from Eq. (23) was used. The sum in Eq. (25) has to be performed over all right states labeled by ν and all left states labeled by μ . No further assumption is made on the nature of these left and right states, i.e. both Bloch states and surface states decaying into the bulk contribute to the current and therefor this formula differs significantly from the Landauer formula.

2.4 Landauer Conductance Versus Bardeen's Tunneling

One might wonder which the difference will be between the results of the Bardeen formula of tunneling and of the Landauer approach. Of course, as stressed in Section 2.3 the results will differ significantly as soon as localized states are present in the vicinity of the barrier region. However, it remains to be clarified what should be expected in the absence of these states. Before the difference between these two transport formulas will be investigated for more realistic systems, it is instructive to look at a very simple model that one can treat analytically.

Figure 7 shows the setup chosen for this simple model, a rectangular barrier of height V_0 and width d between leads in which the electrons are described by free electron wavefunctions. In the Landauer picture of transport, the wavefunction is constructed as

$$\psi(x) = \begin{cases} \frac{1}{k} (\exp(ikx) + r \exp(-ikx)) & x < 0 \text{ in left region} \\ a \exp(-\kappa x) + b \exp(\kappa x) & 0 < x < d \text{ in the barrier} \\ \frac{1}{k} t \exp(ikx) & x > d \text{ in right region.} \end{cases} \quad (26)$$

The decay constant is given by $\kappa = \sqrt{2V_0 - k^2}$, the coefficients a, b and r, t can be determined by wavefunction matching, i.e. by the requirement that the wavefunction and its

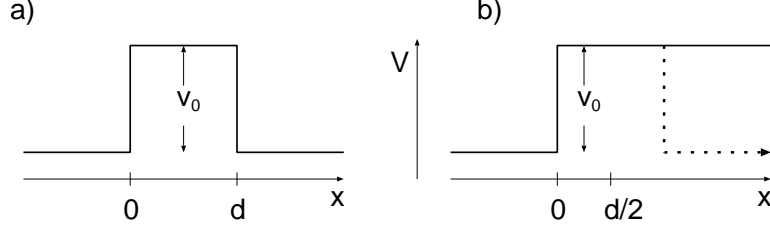


Figure 7. a) Setup of a one-dimensional rectangular barrier used for the analytic evaluation of the Landauer equation. b) Corresponding system for the Bardeen formula. The two sides are separated by extending the barrier to infinity. Only the left system of Bardeen's setup is shown, the right is constructed mirrored.

derivative are continuous at $x = 0$ and $x = d$. The normalization factor $\frac{1}{k}$ has been chosen such that the Bloch states carry unit current as required. Simple algebra reveals the well known formula

$$t = \frac{4i\kappa k e^{-ikd}}{(ik + \kappa)^2 e^{-\kappa d} + (k + i\kappa)^2 e^{\kappa d}}. \quad (27)$$

Inserting this expression into Eq. (10) one obtains the tunneling conductance of this simple barrier in the Landauer approach

$$\begin{aligned} \Gamma_L &= \frac{e^2}{h} \frac{(4\kappa k)^2}{(k^2 + \kappa^2)^2 (1 - e^{-2\kappa d})^2 + (4\kappa k)^2 e^{-2\kappa d}} e^{-2\kappa d} \\ &= \frac{e^2}{h} \left| \frac{4\kappa k}{k^2 + \kappa^2} \right|^2 e^{-2\kappa d} + O(e^{-4\kappa d}). \end{aligned} \quad (28)$$

The second term in this expressions denotes contributions of order $e^{-4\kappa d}$ which can be neglected for any sufficiently thick barrier. This accounts to neglecting the multiple scattering processes in which the electron is scattered multiple times at the interfaces.

If one wants to treat the same system using Bardeen's formula, one has to separate the two systems by extending the barrier to infinity as indicated in Figure 7b). The wavefunctions of the two systems are of course equal except for the transformation $x \leftrightarrow d - x$ and can be written, e.g. for the left side, as

$$\psi_L = \begin{cases} \exp(ikx) + \frac{ik + \kappa}{ik - \kappa} \exp(-ikx) & x \text{ in the leads} \\ \frac{2ik}{ik - \kappa} \exp(-\kappa x) & x \text{ in the barrier.} \end{cases} \quad (29)$$

This formula does not depend on the barrier thickness as it describes completely decoupled systems. The conductance in Bardeen's approach is now given by inserting a separation surface at $x = d/2$ and to evaluate the transition probabilities of Eq. (24) at energy ϵ_0 to obtain

$$\begin{aligned} P_{\text{Bardeen}} &= \frac{\pi \hbar^3}{2m^2} \left| 2 \frac{2ik}{ik - \kappa} e^{-\kappa d/2} \kappa \frac{2ik}{ik - \kappa} e^{-\kappa d/2} \right|^2 \delta(\epsilon - \epsilon_0) \\ &= \frac{\pi \hbar^3}{2m^2} k^2 \left| \frac{4\kappa k}{k^2 + \kappa^2} \right|^2 e^{-2\kappa d} \delta(\epsilon - \epsilon_0). \end{aligned} \quad (30)$$

Finally the summation over all states gives according to Eq. (25)

$$\Gamma_B = \frac{e^2}{h} \left| \frac{4\kappa k}{k^2 + \kappa^2} \right|^2 e^{-2\kappa d}. \quad (31)$$

Hence, both Eqs. (28) and (31) reveal the same conductance in the limit of large κd , i.e. for a small conductance. Figure 8 illustrates this equivalence for a set of parameters typical for an STM setup. It is seen that for barriers thicker than ~ 1 Å Eqs. (31) and (28) give practically identical results.

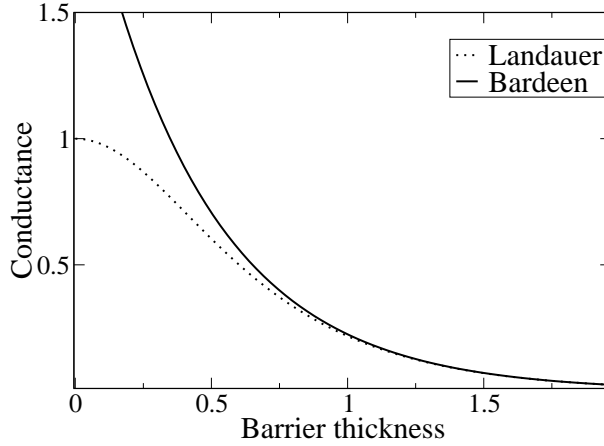


Figure 8. Conductance (in units of the conductance quantum e^2/h) through a rectangular barrier of 5eV barrier height between free electron leads ($\epsilon = 1$ eV) as a function of the barrier thickness (in Å).

2.5 Cu-Vacuum-Cu Tunneling

To demonstrate the two formulas for a more realistic, but still simple case we investigated the electron transmission with normal incidence ($k_{\parallel} = 0$) through a 8.3 Å vacuum barrier separating two Cu(111) surfaces. The calculation was performed within the DFT using the FLAPW-method, as implemented in the embedding version of the FLEUR-code⁹⁻¹¹.

Figure 9 compares the results of the Landauer formula and of Bardeen's approach. In contrast to the simple analytic model discussed before in this setup both localized surface states and delocalized propagating Bloch states can be found. In the energy range below ≈ -0.9 eV a band of Bloch electrons with $k_{\parallel} = 0$ can be found which due to their low energies have very low tunneling probabilities. This transmission probability is described by both formulas and similar to the analytic case discussed before both approaches lead to essentially identical results.

At -0.4 eV, however, a surface state on the Cu(111) surface can be found for $k_{\parallel} = 0$. In the case of the setup required for the Landauer equation these surface states will occur on both sides and hence these states will be split into two levels by hybridization. On the other hand, in Bardeen's approach, because of the complete separation of the two systems

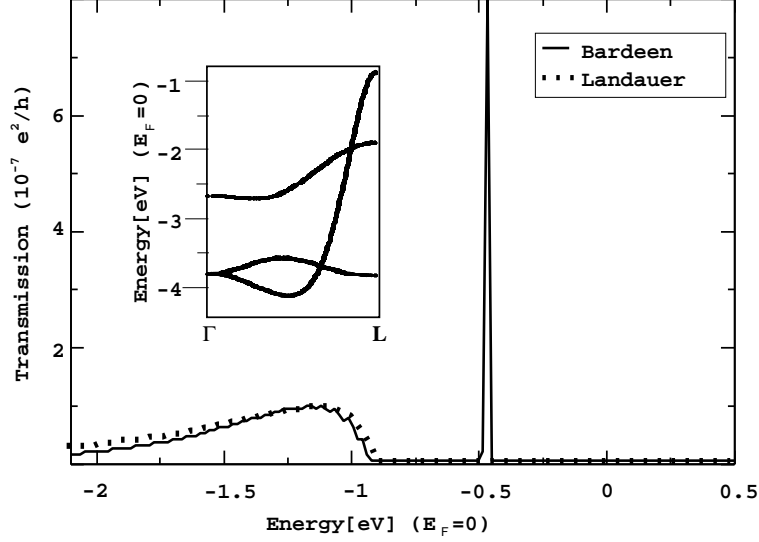


Figure 9. Conductance through a vacuum barrier separating two Cu(111) surfaces. Only electrons with normal incidence are considered. The peak width is due to a finite imaginary part in the energy and its height has been scaled down by a factor of five. The inset shows the normal incidence part of the Cu band structure relevant for the energy window in which transmission from Bloch states can be found from both approaches.

only a single surface state peak occurs and no hybridization splitting is seen in Figure 9. The finite width of this peak is due to the introduction of an imaginary contribution to the energy of $\delta = 0.03$ eV for illustrative purpose only. Such a small broadening of the peak is needed for a numerical integration of the total conductance as well but should not be confused with the modeling of the coupling of the surface state to the leads. For reasonably small imaginary parts the integrated conductance of the peak does not depend on the exact choice of the imaginary energy.

In the case of the setup required for the Landauer equation, one may estimate the electron hopping rate between the two levels from the energy splitting. Nevertheless, these states do not contribute to the conductance in the Landauer equation as no Bloch states are present in the leads at this energy. Bardeen's equation on the other hand contains a strong contribution from the surface state. In this very artificial situation with the same surface state on both sides of the barrier this effect can only be seen at exactly zero bias. However, the arguments presented here are equally valid for situations in which one encounters a localized state at one side of the barrier and a continuum at the other.

In order for electron hopping between two surface states to be measured as the current, they must be coupled to extended states in both leads, for example, via impurity-induced random potentials, electron-phonon interaction,¹² and electron-electron scattering.¹³ Applying Bardeen's equation to the surface-state conductance assumes implicitly that the transition between two localized states on both sides is the rate-limiting process.

The present results clearly indicate the limits of validity of the two different approaches. While the Landauer formula will be suitable in cases in which a relatively high conductance

is obtained by the coupling of Bloch states, it must be applied with care for tunneling setups in which states localized at interfaces might play a crucial role. On the other hand the Bardeen approach to tunneling is only suitable for exactly these situations and requires a high barrier leading to a low conductance.

3 DFT Calculations of Transport Setups

The most challenging problem when evaluating most of these transport equations starting from a DFT single-particle description is the particular geometry of the system. While most computer codes are build to treat either (i) periodic systems making use the periodic boundary conditions and the Bloch construction of the states or (ii) finite systems like molecules or clusters with vacuum boundary conditions, in a transport system one has to deal with an open system. At both sides of the region of interest in which the quantum scattering on some potential perturbation takes place, semi-infinite leads are attached. The system has no periodicity in this direction of space and solving the Kohn-Sham equation within the scattering volume will only be possible by taking the appropriate boundary condition into account which ensures that the effects of the semi-infinite leads are treated correctly.

To formalize the task, we want to treat this scattering volume only, i.e. we want to deal with the Kohn-Sham Hamiltonian limited to this finite volume H_Ω . Of course we cannot simply consider the equation

$$H_\Omega \psi_\Omega = \epsilon \psi_\Omega \quad (32)$$

since such a simple approach will completely miss out the effect of the attached leads. In fact the way we wrote Eq. (32) it is not even well defined. The Hamiltonian of the total system can not be written as

$$H_\Omega + H_L + H_R \quad (33)$$

containing contributions from the left H_L and the right H_R lead as well as the central region as such a description does not contain the coupling between these volumes. Hence the Kohn-Sham Hamiltonian can not simply be restricted to the center region without specifying the boundary condition imposed at the boundary of this region or equivalently, without considering the coupling to the leads and the properties of this leads.

In order to understand a scheme suitable to achieve the desired treatment of the barrier volume only, we write the total Hamiltonian of the system in matrix form

$$H = \begin{pmatrix} H_L & V_{L\Omega} & 0 \\ V_{\Omega L} & H_\Omega & V_{\Omega R} \\ 0 & V_{R\Omega} & H_R \end{pmatrix}, \quad (34)$$

where $V_{L\Omega}$ and $V_{R\Omega}$ denote the coupling of the left and right lead to the central scattering region. As the Kohn-Sham Hamiltonian is local no direct coupling between the leads needs to be considered and one obtains Eq. (34) by using any sufficiently localized basis set. Starting from this Hamiltonian the Green function of the scattering volume G_Ω can be obtained. The single-particle Green function is formally defined by

$$(H(\vec{r}) - \epsilon) G(\vec{r}, \vec{r}'; \epsilon) = \delta(\vec{r} - \vec{r}'), \quad (35)$$

or more concise in matrix notation

$$(H - \epsilon) G(\epsilon) = 1. \quad (36)$$

Our goal is the calculation of the Green function for the total Hamiltonian restricted to the scattering volume Ω . In analogy to the partitioning of the Hamiltonian we can write

$$\begin{pmatrix} H_L - \epsilon & V_{L\Omega} & 0 \\ V_{\Omega L} & H_\Omega - \epsilon & V_{\Omega R} \\ 0 & V_{R\Omega} & H_R - \epsilon \end{pmatrix} \begin{pmatrix} G_L & G_{L\Omega} & G_{LR} \\ G_{\Omega L} & G_\Omega & G_{\Omega R} \\ G_{RL} & G_{R\Omega} & G_R \end{pmatrix} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (37)$$

Hence, the Green function G_Ω is defined by

$$V_{\Omega L} G_{L\Omega} + (H_\Omega - \epsilon) G_\Omega + V_{\Omega R} G_{R\Omega} = 1. \quad (38)$$

From the off-diagonal elements of Eq. (37) one obtains ($X = L$ or $X = R$)

$$\begin{aligned} G_{X\Omega} &= -(H_X - \epsilon)^{-1} V_{X\Omega} G_\Omega \\ &= -G_X^0 V_{X\Omega} G_\Omega, \end{aligned} \quad (39)$$

where the Green function $G_X^0 = (H_X - \epsilon)^{-1}$ of the isolated leads has been introduced. consequently one can rewrite Eq. (38) into

$$(H_\Omega - \epsilon - \Sigma_L - \Sigma_R) G_\Omega = 1. \quad (40)$$

This demonstrates that one can calculate the Green function in the scattering volume G_Ω by adding two additional terms $\Sigma_{L/R}$ to the Hamiltonian which take into account the effect of the attached semi-infinite leads. These so-called self-energies are given by

$$\Sigma_X = V_{\Omega X} (H_X - \epsilon)^{-1} V_{X\Omega} = V_{\Omega X} G_X^0 V_{X\Omega}. \quad (41)$$

and play a crucial role in the *ab initio* calculation of scattering setups. By means of the self-energy the coupling to the leads is described. Consequently, both the interaction between the leads and the scattering volume $V_{X\Omega}$ and the electronic structure of the leads H_X enter in its definition. Since these Σ are energy dependent, one can not easily cast Eq. (38) into an eigenvalue problem to obtain the single-electron wavefunctions but instead must express the quantities of interest in terms of the Green function.

The linear response conductivity of the Kohn-Sham system has been expressed by many authors in terms of the Green function. This approach yields

$$\Gamma = \frac{e^2}{h} \text{Tr} (G_\Omega \Im \Sigma_R G_\Omega^* \Im \Sigma_L). \quad (42)$$

One can prove^{14,9} that this expression is equivalent to the Landauer formula as given in Eq. (10). We will not repeat this derivation here but instead give some handwaving arguments which create understanding by analogies in the interpretation of the different constituents of the expression:

- As one can see from Eq. (41) the imaginary part of the self-energies of the two leads is due to the Green function of the leads G_X^0 . Hence the imaginary part of the self energy contains the information on the Bloch states in the leads and projects out the propagating states in the trace performed in Eq. (42) in analogy to the sum over asymptotic Bloch states in the original Landauer equation (Eq. (10)).

- As the self-energy describes the coupling to the leads they are highly localized at the surface between the scattering volume of the leads and in products of the form $\Im \Sigma_L G \Im \Sigma_R$ the two spacial arguments of the Green are basically placed on the two opposite sides of the scattering volume. Hence this Green function basically corresponds to the transmission coefficient in the original Landauer equation.

3.1 Non-Equilibrium Calculation

The formalism we discussed so far is restricted to the treatment of electrical conductivity in the linear response regime valid for the limit of zero bias voltage. This limit is definitely appropriate for systems with nearly perfect transmission. If there would be very little scattering of the incoming electrons, i.e. if the incoming Kohn-States do not couple to reflected states but are perfectly transmitted, it would be impossible to apply a large bias voltage. This can be understood very simply from Ohm's law which tells you that you can not have a potential drop if you have no resistance. Hence, in this highly conducting case the zero bias treatment is the only reasonable description as long as one does not go beyond our model of ballistic transport without scattering processes making the DFT description invalid.

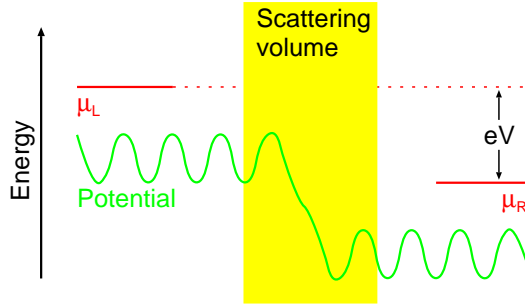


Figure 10. Potential and local Fermi-levels in a system out of equilibrium. The bias applied between the two leads gives rise to a potential drop which is confined to the scattering volume so that the leads stay in local equilibrium.

The situation is different in the case of a strong reflection of the electrons within the scattering volume. In this case a substantial potential drop occurs within the scattering volume and this effect can be treated. In the limit of very little transmission like in a tunneling setup, one can safely assume that the total resistance measured in an experiment is actual due to the tunnel barrier, i.e. to the effect of the scattering volume. In this case the potential is flat in the leads, and all the potential drop will be across the barrier.

The basic assumption one makes when calculating a conducting junction under non-equilibrium conditions is the neglect of the effect of the applied bias on the leads. This includes the assumption that the potential in the leads is “flat” i.e. the leads are assumed to have the bulk potential. Additionally, no effect of the current in the leads is considered, the electronic structure of the leads is unchanged from the situation of the bulk. These assumptions of course require that the current is reasonably small.

We will consider the bias voltage V as being applied to the left lead changing the local potential in the lead

$$V_L \rightarrow V_L + eV. \quad (43)$$

As the leads are assumed to be in a local equilibrium one can assign a local chemical potential $\mu_{(L/R)}$ to both leads. These different chemical potential levels are shifted according to the applied bias voltage

$$\mu_L = \mu_R + eV. \quad (44)$$

The basic task in the self-consistent DFT cycle for the system in this non-equilibrium situation is the calculation of the charge density in the scattering volume. This charge density can be viewed as being composed out of the charge due to electrons incident from the left lead N_L and the charge due to electrons incident from the right N_R ^a

$$N = N_L + N_R. \quad (45)$$

Due to the different Fermi levels in the leads two different Fermi distribution functions f have to be considered in the evaluation of these charges

$$N_L = \frac{1}{\pi} \int_{-\infty}^{\infty} f_L(\epsilon) A_L(\epsilon) d\epsilon \quad \text{and} \quad N_R = \frac{1}{\pi} \int_{-\infty}^{\infty} f_R(\epsilon) A_R(\epsilon) d\epsilon, \quad (46)$$

where $f_{L/R}(\epsilon) = f(\epsilon - \mu_{(L/R)})$ and $A_{L/R}$ denotes the spectral function due the left and right incoming states. This left and right spectral functions are key quantities in non-equilibrium calculations. They are given in terms of Kohn-Sham states $\psi_{L/R}$ which are incoming from the respective lead ^b

$$A_{L/R}(\epsilon) = \pi \sum_i \psi_{i;L/R}^* \psi_{i;L/R} \delta(\epsilon_i - \epsilon). \quad (47)$$

The sum has to be performed over all possible incoming electron states. While some codes exist that use such an expression of the spectral function in terms of the wavefunctions most approaches employ a reformulation of the spectral function in terms of the Green function of the scattering volume and the self-energies of the leads.

$$A_{L/R} = G^* \Im \Sigma_{L/R} G. \quad (48)$$

This equation can be derived from the definition of the spectral function as the imaginary part of the Green function. Hence, assuming that the spectral function can be separated into a left/right part one can write

$$A_L + A_R = \frac{-i}{2} (G - G^*). \quad (49)$$

The Green function of the scattering volume is given in terms of the Hamiltonian and the self-energies of the leads (Eq. (38))

$$G^{-1} = \epsilon - H - \Sigma_L - \Sigma_R \quad (50)$$

^a Actually, this statement is not valid if states exist which are localized within the scattering volume and which decay exponentially into the leads. Such states impose a formidable problem if they are energetically located between the two Fermi levels. In this case the occupation of the localized state is not defined without considering further effects beyond the simple DFT approach.

^b These states are usually called scattering states.

and

$$G^{*-1} = \epsilon - H - \Sigma_L^* - \Sigma_R^*, \quad (51)$$

as the Hamiltonian is Hermitian. Subtracting Eqs. (50) and (51) and multiplying with G^* from the left and G from the right one obtains

$$\frac{-i}{2}(G - G^*) = G^* \Im \Sigma_L G + G^* \Im \Sigma_R G \quad (52)$$

from which one can easily identify Eq. (48).

We will finalize the discussion of this expression of the left/right spectral function with some comments:

- While we only presented a rough and handwaving derivation of these expressions, one can also obtain them most rigorously within the non-equilibrium Keldysh Green function technique. Such a derivation also allows to systematically improve the level of approximation by including further interactions in a perturbative treatment.
- The two expressions in terms of the Green functions and in terms of the wavefunctions are equivalent. However, in practical calculations the Green function formalism is more widely used.
- The presence of localized states in the scattering volume introduces a significant problem in non-equilibrium calculations. Since these states do not couple to the Bloch states of the left or right leads it is unclear how the occupancy of such states has to be chosen as soon as their energy will be between the two Fermi levels. In reality, the occupancy of these states is determined by the scattering processes beyond the DFT which provide a small coupling to the leads. However, since these processes are not modeled in the simple non-interacting DFT calculation, additional assumptions must be introduced to deal with localized states.
- The decomposition of the spectral function into a right and left part allows to interpret the Landauer equation in a slightly different way. Comparing Eqs. (42) and (48) one realizes that

$$\Gamma = \frac{e^2}{h} \text{Tr} (A_L \Im \Sigma_R) = \frac{e^2}{h} \text{Tr} (A_R \Im \Sigma_L). \quad (53)$$

Hence, the Landauer conductance is given by the projection of the left spectral function on the Bloch states of the right lead or vice versa.

3.2 Typical Flow of a Non-Equilibrium Calculation

We will finalize our discussion by summarizing the different tasks one has to perform in order to put the non-equilibrium scheme into practice. These tasks are summarized in Figure 11 as well. One of the most crucial parts is the calculation of the self-energy of the leads. When using the definition of Eq. (41) the most difficult problem in this step is the calculation of the Green function of the isolated lead. While the Green function of the infinite lead is relatively easy to obtain, the calculation of the Green function $G_{L/R}^O$ of the isolated, semi-infinite leads imposes a substantial problem due to the breaking of

translational symmetry. The missing hopping to the central scattering volume make the first layers of the lead substantially different from the bulk, while – at the same time – the potential should remain unchanged. Different approaches have been developed to solve this problem, most of which employ some kind of iterative calculation of the Green function or the self energy. In many codes this calculation of the self-energies is a time-consuming and difficult task.

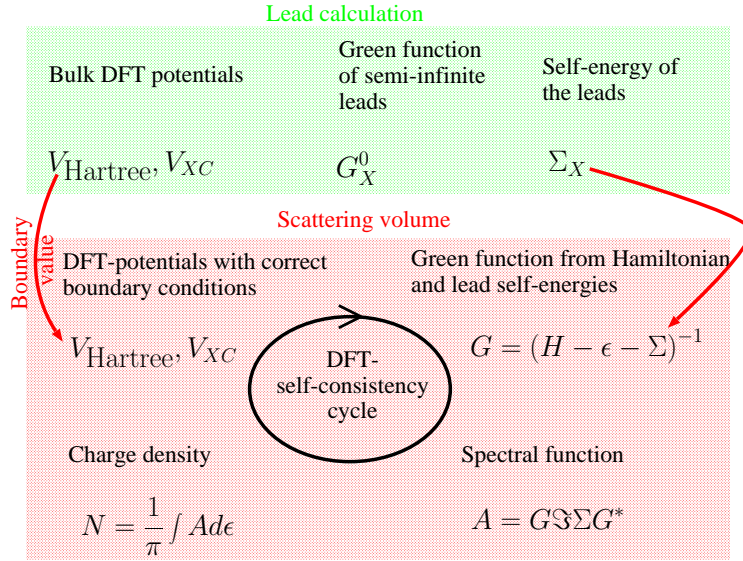


Figure 11. Flow of a self-consistent calculation using the non-equilibrium Green function formalism.

Once the self-energy of the leads is known, the self-consistent calculation of the charge density in the scattering volume can be started. The two major steps here are the calculation of the Hartree potential in the scattering volume and the subsequent calculation of the left and right spectral functions.

The applied bias enters the calculation of the Hartree potential. At the boundaries of the central volume, the Hartree potential has to fulfill a Dirichlet boundary condition imposed by the bulk Hartree potential of the leads. The bias shifts this potential on one side of the leads effectively imposing an electric field into the scattering volume. Obviously, the Hartree potential depends on the charge density within the scattering volume and hence this calculation is the starting point of a DFT self-consistency cycle.

The calculation of the Green function of the scattering volume including the self-energies of the lead is the next important task. Very often this is a very time consuming and memory intensive part as a possibly very large Hamiltonian of the scattering volume has to be inverted (see Eq. (38)) for a huge set of different energies and possibly different \vec{k}_{\parallel} -values in a periodic junction. The Green function of the scattering volume then allows the calculation of the left and right spectral functions in accordance with Eq. (48). The DFT self-consistency cycle will ensure that the charge redistribution due to the applied

bias is taken into account properly. For example, screening charges will be induced at the interface such that the metallic leads become free of the external electric field and the applied bias voltage, which will lead to a simply linear potential drop across all the scattering volume without considering the self-consistent redistribution of the charge, will be confined to a smaller volume which includes only the scattering potential.

4 Summary

Electronic structure calculations on the basis of the density functional theory allow to investigate the effect of details of the atomic structures on the transport in nanostructures. We discussed the approximations most frequently used in these calculations, the Landauer formula and Bardeen's tunneling approach and stressed their similarities as well as the differences in the treatment of the tunneling conductance. Finally, we introduced the basic ideas used to treat systems out of equilibrium in which a finite bias voltage is applied across a scattering region. The key formulas of the non-equilibrium Green function technique applied to the non-interacting DFT system were introduced and interpreted.

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